# Supporting Information

## Mechanical modulation of 2D transition metal dichalcogenide alloys

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### Young's moduli and pretension measurements

Additional Young's moduli and pretension measurements as a function of the atomic composition, x, are shown in Figures S1 and S2, respectively. In Figure S3 we present the Young's moduli and pretension of WS<sub>2</sub> and MoS<sub>2</sub>.

#### Additional Raman and photoluminescence measurements

Raman measurements of 19 nm thick nano-drumheads having different W concentrations are shown in Figure S4. PL measurements of 19 nm and 28 nm thick nano-rumheads are shown in Figure S5.

### Additional atomistic simulation results

More information obtained from the atomistic simulations is shown in Figures S6 and S7.



Fig. S1. Young's moduli of nano-drumheads of different thicknesses (noted on the graphs) as a function of W concentration, x. Trendlines are shown as dashed lines.



Fig. S2. Pretension of nano-drumheads of different thicknesses (noted on the graphs) as a function of W concentration, x.



Fig. S3. Younng's moduli and pretension of  $WS_2$  and  $MoS_2$  nano-drumheads as a function of the thicknesses.



Fig. S4. Raman measurements of 19 nm thick nano-drumheads of different W compositions.



Fig. S5. PL measurements of (a) 19 nm and (b) 28 nm thick nano-drumheads of different W compositions.



Fig. S6. Atomistic simulation results: the stress distribution of different alloys in atomistic simulations. Left column – The atomic distribution of the central metallic (W/Mo) layer. Right column – The atomic local stress times the local atomic volume.



Fig. S7. The third-order elastic modulus as a function of the W concentration obtained from the atomistic simulations.